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DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

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U.S. APPLICATION NO. (if known, see 37 CFR 1.5)

09/890296

INTERNATIONAL APPLICATION NO.  
PCT/JP00/08458INTERNATIONAL FILING DATE  
29 November 2000 (29.11.00)PRIORITY DATE CLAIMED  
03 December 1999 (03.12.99)

TITLE OF INVENTION

LIQUID PURIFICATION APPARATUS

APPLICANT(S) FOR DO/EO/US

OKAUE, Kimihiko

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☐ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
  - b. ☒ has been communicated by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
  - a. ☒ is attached hereto.
  - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
  - b. ☒ have been communicated by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☐ have not been made and will not be made.
8. ☒ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

## Items 11 to 20 below concern document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☐ A **FIRST** preliminary amendment.
14. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information:  
International Search Report

U.S. APPLICATION NO. (if known, see 37 CFR 1.5) <b>09/890296</b>		INTERNATIONAL APPLICATION NO. PCT/JP00/08458		ATTORNEY'S DOCKET NUMBER 33707	
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21. <input checked="" type="checkbox"/> The following fees are submitted: <b>BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):</b> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO..... \$1000.00  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... \$860.00  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$710.00  International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$690.00  International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$100.00  <b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				<b>CALCULATIONS PTO USE ONLY</b>          																															
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$ 860.00																															
<table border="1" style="width:100%; border-collapse: collapse;"> <tr> <th style="width:15%;">CLAIMS</th> <th style="width:15%;">NUMBER FILED</th> <th style="width:15%;">NUMBER EXTRA</th> <th style="width:15%;">RATE</th> <th style="width:15%;">\$</th> <th style="width:15%;"></th> </tr> <tr> <td>Total claims</td> <td>12 - 20 =</td> <td>0</td> <td>x \$18.00</td> <td>\$</td> <td></td> </tr> <tr> <td>Independent claims</td> <td>6 - 3 =</td> <td>3</td> <td>x \$80.00</td> <td>\$</td> <td>240.00</td> </tr> <tr> <td colspan="3">MULTIPLE DEPENDENT CLAIM(S) (if applicable)</td> <td>+ \$270.00</td> <td>\$</td> <td>270.00</td> </tr> <tr> <td colspan="4" style="text-align: right;"><b>TOTAL OF ABOVE CALCULATIONS =</b></td> <td>\$</td> <td>1,370.00</td> </tr> </table>				CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$		Total claims	12 - 20 =	0	x \$18.00	\$		Independent claims	6 - 3 =	3	x \$80.00	\$	240.00	MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$270.00	\$	270.00	<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$	1,370.00		
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<input checked="" type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$ 685.00																															
<b>SUBTOTAL =</b>				\$ 685.00																															
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$																															
<b>TOTAL NATIONAL FEE =</b>				\$ 685.00																															
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +				\$																															
<b>TOTAL FEES ENCLOSED =</b>				\$ 685.00																															
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a. ☒ A check in the amount of \$ 685.00 to cover the above fees is enclosed.

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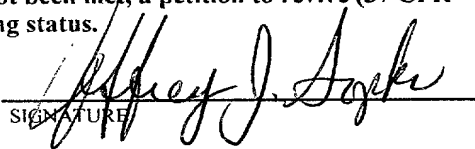
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 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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"EXPRESS MAIL" MAILING LABEL CERTIFICATE

Re: U.S. PCT-based Patent Application for  
"LIQUID PURIFICATION APPARATUS"  
International Application No.: PCT/JP00/08458  
International Filing Date: November 29, 2000  
Attorney's Docket No.: 33707

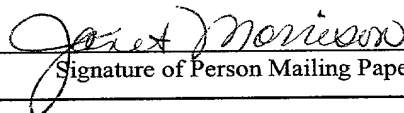
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09/890296 07/27/01

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JC18 Rec'd PCT/PTO 27 JUL 2001

- 1 -

## DESCRIPTION

### LIQUID PURIFICATION APPARATUS

#### TECHNICAL FIELD

The present invention relates to a liquid purification apparatus for purifying a wastewater, i.e., a liquid to be treated containing at least one of minute particles (for example, minute particles of dioxins, bacteria and the like), metal ions, heavy metal ions and the like, such as, for example, a wastewater from an incinerator.

#### BACKGROUND ART

Conventionally, filtration of a wastewater containing very fine minute particles such as those of 0.1  $\mu$ m or the like could not be effected even with ceramic filters having many minute apertures, and no other measure has been known except removal with hollow yarns. In addition, running costs required for an operation for solving clogging of ceramic filters are considerably high.

Therefore, in the conventional cases, minute particles were aggregated and precipitated by adding an aluminum or magnesium aggregation agent to the waste fluid, and the precipitated aggregates were removed by filtering with a press filter.

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In addition, no other measure has been known for removal of heavy metals in discharged water except removal with a chelating agent effected separately from the above operation or removal with an ion-exchange resin.

#### PROBLEM TO BE SOLVED BY THE INVENTION

Measures as described above have been applied in the conventional apparatuses and methods, and removal of very fine minute particles has been made by aggregation and precipitation of the minute particles together with an aggregation agent through addition of the aggregation agent. Therefore, the aggregation agent should be added in a large amount and costs for the aggregation-precipitation product itself as well as its waste treatment could not be ignored. In addition, a large amount of precipitates were produced by the addition of a large amount of the aggregation agent, and hence the operation for filtration thereof under pressure with a press filter became enormous and costs therefore were remarkable.

Moreover, the removal of metal ions and heavy metal ions could only be performed with a chelate agent or an ion-exchange resin; this required an enormous expenses for the initial costs and the running costs and a problem occurred in the regeneration of a chelate agent that a large amount of a concentrated wastewater containing

heavy metal ions was produced.

#### DISCLOSURE OF INVENTION

The invention has been made for solving the problems in conventional measures as described above.

The invention is made such that the purification is attained by placing an adsorbent comprising at least basic magnesium sulfate and magnesium hydroxide between a first and a second filter layers, passing a liquid to be treated through the adsorbent, thereby, causing aggregation of minute particles in the liquid to be treated so that a large mass is formed and then trapping it by the second filter layer.

This allows removal of the minute particles in the liquid to be purified without using a large amount of an aggregation agent, and the minute particles to be removed could be trapped and removed with a filter having openings about 100 times as large as the size of the minute particles to be removed.

In the invention, "purification" does not refer to a simple filtration with a filter, such as a trapping, in a space, of a particulate substance contained in a liquid to be treated by passing the liquid through the space having a given size, but refers to a trapping of a targeted substance in the above described space having a given size after aggregating the substance to form a mass having a certain

dimension.

In another feature of the invention, a purification apparatus for a liquid to be treated is provided, such that metal ions are trapped by placing an adsorbent comprising at least basic magnesium sulfate and magnesium hydroxide between a first and a second filter layers, passing the liquid to be treated containing the metal ions through the adsorbent, thereby causing binding of the metal ions, particularly heavy metal ions, in the liquid to be treated to a hydroxyl group in an adsorbent resulting in flocculation, causing aggregation of flocculated particles so that a large mass is formed, and then trapping it by the second filter layer.

This allows removal of the metal ions or the heavy metal ions in the liquid to be treated without using a chelate agent or an aggregation agent.

Another feature of the invention is such that metal ions is trapped by placing an adsorbent comprising at least basic magnesium sulfate and magnesium hydroxide between a first and a second filter layers, electrically charging a direct current by electrode equipments, causing generation of OH ions in the liquid to be treated, causing binding of the OH ions and a hydroxyl group in the above described adsorbent to the metal or heavy metal ions in the liquid to be treated resulting in flocculation, causing aggregation of flocculated particles so that a large mass

is formed, and then trapping it by the second filter layer. This allows removal of the metal ions or the heavy metal ions in the liquid to be treated with a low-cost apparatus without using a chelate agent or an aggregation agent.

Another feature of the invention is designed such that metal ions are trapped by placing an adsorbent comprising a powdery cellulose between a first and a second filter layers, electrically charging a direct current by electrode equipments, causing generation of OH ions in the liquid to be treated, causing binding of this OH ions and a hydroxyl group in the above described adsorbent to the metal or heavy metal ions in the liquid to be treated resulting in flocculation, and then trapping flocculated particles by the adsorbent and the second filter layer.

This allows removal of the metal ions or the heavy metal ions in the liquid to be treated with a low-cost apparatus without using a chelate agent or an aggregation agent.

#### BRIEF DESCRIPTION OF DRAWINGS

Fig. 1 is a side sectional view showing an example of a purification apparatus for a liquid to be treated according to the invention.

Fig 2 is a graph of a characteristic curve for the potential energy.

Fig. 3 is a graph showing a condition of metal



contents in a waste fluid from silicon electrolysis before purification obtained by an X-ray microanalyzer.

Fig. 4 is a graph showing a condition of metal contents after 3 times of repeated purification frequency with the example in Fig. 1 obtained by an X-ray microanalyzer.

Fig. 5 is a graph showing a condition of metal contents after 20 times of repeated purification frequency with the example in Fig. 1 obtained by an X-ray microanalyzer.

Fig. 6 is a side sectional view showing another example of a purification apparatus for a liquid to be treated according to the invention.

Fig. 7 is a side sectional view showing a still another example of a purification apparatus for a liquid to be treated according to the invention.

Fig. 8 is a graph showing a condition of metal contents in a waste fluid from silicon electrolysis before purification obtained by an X-ray microanalyzer.

Fig. 9 is a graph showing a condition of metal contents after 3 times of repeated purification frequency with the example in Fig. 7 obtained by an X-ray microanalyzer.

Fig. 10 is a graph showing a condition of metal contents after 20 times of repeated purification frequency with the example in Fig. 7 obtained by an X-ray

microanalyzer.

Fig. 11 is an expanded photograph in 30 magnifications of an adsorbent composed of 3 substances shown by Chemical Formula 1, Chemical Formula 2 and Chemical formula 3 obtained by a scanning electronic microscope.

Fig. 12 is an expanded photograph in 300 magnifications of an adsorbent composed of 3 substances shown by Chemical Formula 1, Chemical Formula 2 and Chemical formula 3 obtained by a scanning electronic microscope.

#### BEST MODE FOR CARRYING OUT THE INVENTION

##### [First Example]

An example of the invention is described below with reference to drawings. Fig. 1 is a side sectional view showing an example of a purification apparatus according to the invention.

In the drawing, a waste fluid containing, for example, minute particles or heavy metal ions is charged under pressure by a pump from an inlet 2 for a liquid to be treated of a container 1. First and second filter layers 3 and 4 are placed in the container 1 and an adsorbent 5 is filled between the first and second filter layers 3 and 4. A built-up body with a cylindrical resin framework of polypropylene having a height of 500 mm and a diameter of 120 mm composed integrally of the first and second filter layer 3 and 4 and the adsorbent 5 constitutes an adsorbent

cassette. The cassette was installed in the container 1 in the form of a cylinder, made of stainless steel, having a height of 750 mm and a diameter of 150 mm as shown in Fig. 1.

The adsorbent 5 filled in said adsorbent cassette consists preferably of:

94% or more of a basic magnesium sulfate represented by:  $\text{MgSO}_4 \cdot 5\text{Mg}(\text{OH})_2 \cdot 3\text{H}_2\text{O}$  [Chemical Formula 1]

6% or less of magnesium hydroxide represented by:  $\text{Mg}(\text{OH})_2$  [Chemical Formula 2] and

0.5% or less of magnesium sulfate represented by:  $\text{MgSO}_4$  [Chemical Formula 3] ;

and its geometry is powdery or granular or particulate form, constituted of gathered crystalline fibers. Expanded photographs by a scanning electronic microscope are shown in Fig. 11 and Fig. 12. Fig. 11 and Fig. 12 are expanded photographs in 30 magnifications and 300 magnifications, respectively, of the adsorbent 5 composed of three substances shown by Chemical Formulae 1, 2 and 3.

The ratio of the contained components is not limited to the above-mentioned example, and may be anyone within a range of:

$2\% \leq \text{MgSO}_4 \cdot 5\text{Mg}(\text{OH})_2 \cdot 3\text{H}_2\text{O} \leq 98\%$  [Chemical Formula 4] and

$2\% \geq \text{Mg}(\text{OH})_2 \geq 98\%$  [Chemical Formula 5] .

When the amount of basic magnesium sulfate and that of magnesium hydroxide are less than or more than the above-mentioned range, they do not have a desired geometry (a geometry of a minute pill-like structure shown by the photographs of scanning electronic microscope in Fig. 10 and Fig. 11).

That is, in the layer of adsorbent 5 sandwiched between the first and second filter layers 3 and 4, the concentration of metal ions becomes somewhat higher than that of metal ions in the liquid to be treated by the adsorbent 5. This fact can be explained by the Schulze Hardy's law for some kinds of liquids to be treated. However, for some other kinds of liquids to be treated, that fact can not necessary be explained. However, according to experiments, for all the substances of non-acidic liquids to be treated, the liquid purification according to the invention was effective.

Next, action in the first example described above is illustrated with Fig. 2.

Fig. 2 is a characteristic curve for the potential energy; the particle distance  $L$  between minute particles A and B is plotted along the abscissa axis and the repulsion force  $P_1$  and the absorption force  $P_2$  are plotted along the ordinate axis.

When a liquid to be treated containing heavy metal

ions are charged through the inlet 2 for the liquid to be treated into the container 1 in Fig. 1 under pressure by a pump, the waste fluid flows through the first filter layer 3 to the adsorbent 5. Because the adsorbent 5 has a powdery or granular or particulate constitution, the waste fluid passes through in-between spaces. The mutual distance L between the minute particles A and B in the liquid to be treated shown in Fig. 2 changes from distance  $L_1$  to distance  $L_2$  or less; an attractive force  $P_2$  acts between the minute particles A and B in the liquid to be treated so that they aggregate together forming large masses of about 300 times in size. This has been confirmed by collaboration with Osaka Municipal Technical Research Institute.

The minute particles in the form of large masses are trapped and removed by the second filter layer 4 (placed downstream of the adsorbent 5 in the flowing direction of the liquid to be treated) having sufficiently rough openings as compared with individual minute particles.

For example, even the minute particles are of  $0.1 \mu\text{m}$  and could conventionally be trapped only by hollow yarns, they can be removed with a filter of  $10 \mu\text{m}$  used as the second filter layer.

In the example, a filter paper having a mesh size of  $100 \mu\text{m}$  was used as the first filter layer 3. The mesh size may be anyone that has openings finer than the powdery or granular or particulate adsorbent 5.

In addition, when the adsorbent 5 is in a powdery form, there is a possibility of causing clogging in spaces, because in-between spaces between powders are narrow. In this case, the clogging may be prevented by broadening the in-between spaces by using as spacers, for example, an activated carbon in particulate form or the like having a specific gravity close to that of the adsorbent 5. When spaces are broadened by inserting spacers in this manner, action of the adsorbent described below has not been adversely affected.

Silica which causes clogging of reverse osmotic membranes has conventionally been considered irremovable. However, by using the example having a constitution of Fig. 1, it can be removed to an extent in which the clogging of reverse osmotic membranes can sufficiently be prevented, as shown in Table 1. In this case, well water (the well water has generally a high content of silica) was used as the liquid to be purified (liquid to be treated), and a cartridge filter of 10  $\mu$ m was used as the second filter layer 4. Purification frequency in Table 1 refers to a frequency of passing with repeated circulation of the liquid to be treated through the purification apparatus having the constitution shown in Fig. 1.

[Table 1]

Purification Frequency	Well water	Once	3 times	5 times	10 times	20 times
Silica (mg/l)	34.4	25.0	21.0	16.3	15.4	7.8

In the desalinization of seawater, in order to prolong the period before causing clogging of reverse osmotic membrane, various measures have been studied for suppressing the FI value (Fouling Index or Silt Density Index) as an index to indicate the degree of contamination to 3 or below. However, it has been considered that the FI value cannot be lowered to 3 or below, up to the present.

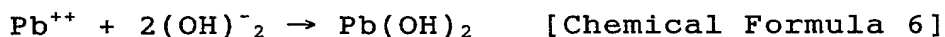
By using the example in Fig.1, however, the FI value could easily be lowered to 3 or below as shown in Table 2.

In this case, a cartridge filter of 10  $\mu$ m was used as the second filter layer 4. As method of measurement therein, a method of December 1987, established by Nitto Electric Chemical Co., Ltd., Japan, was used.

[Table 2]

Purification frequency	Seawater	5 times	10 times
FI value	4.31	2.62	2.47

In addition, when heavy metal ion, for example, lead in the liquid to be treated passes through an in-between spaces between the adsorbents 5 in powdery or granular or particulate form, lead ions is bound to a hydroxyl group in the adsorbent 5 causing a reaction shown in the following [Chemical Formula 6]:



As shown by the [Chemical Formula 6], lead ions  $\text{Pb}^{++}$  is converted to  $\text{Pb}(\text{OH})_2$  forming flocks; and the flocks aggregate while passing through the adsorbent 5 forming masses of about 300 times in size, and is trapped by the second filter layer 4.

Table 3 shows result measured using atomic absorption analysis in washing of worker's uniforms with lead powders adhered in a plant; the result shows degree of lead removal in washing wastewater. In this case, a cartridge filter of 10  $\mu\text{m}$  was used as the second filter layer 4.

[Table 3]

Purification Frequency	Wastewater	Once	3 times	5 times
Lead content (ppm)	0.696	0.015	0.007	0.004

As shown in Table 3, lead minute particles and lead ions, which have conventionally been considered removable only with a chelate agent or a high molecule aggregation agent, in the wastewater was able to be removed by the apparatus of the invention; and the costs therefor were 1/10 or less in the initial costs and 1/20 or less in the running costs as compared with the conventional one. In addition, in the case of the apparatus of the invention,



a space for placing the installation was about 1/30 as compared with the above-described conventional method.

Next, an example for removing heavy metal ions in a silicon electrolysis waste fluid from a factory for processing silicon material for semiconductor element in Example 1 of Fig. 1 is described.

Fig. 3, Fig. 4 and Fig. 5 show graphs of conditions of metal contents obtained by an X-ray microanalyzer using the apparatus of the example in Fig. 1.

Fig. 3 shows metal contents in a waste fluid from silicon electrolysis before purification; Fig. 4 shows metal contents in the purification liquid after 3 times of purification frequency, being purified with a cartridge filter of 10  $\mu$ m as the second filter layer 4, and; Fig. 5 shows metal contents in the purified liquid after 20 times of circulated purification frequency, being purified with a cartridge filter of 10  $\mu$ m as the second filter layer 4, respectively.

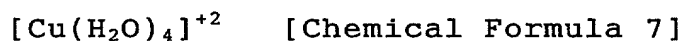
The data are summarized in Table 4.

[Table 4]

Metal	Co	Fe	Cu	Si
Waste fluid (mg/l)	7.76	5.20	9.08	30.17
3 times (mg/l)	0.46	1.12	5.58	0.00
20 times (mg/l)	0.10	0.74	5.09	0.00

As shown in Table 4, the heavy metals were removed to an almost satisfactory degree.

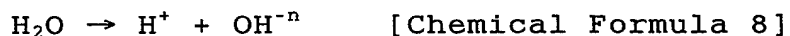
As an exception, as shown in [Chemical Formula 7], copper ions exist in water in a state in which it can hardly bind to hydroxyl group. Therefore, removal thereof was considerably difficult.



[Second Example]

Fig. 6 is a side sectional view showing the second example of the invention, and made in the same size and material as that in Fig. 1. In Fig. 6, however, an electrode 6 is composed, for example, in a network form; and, for example, a direct voltage of +2 V is charged using the container 1 as the earth electrode; the electrode 6 and the container 1 constitute electrode equipments. As the adsorbent 5, the same material as that in the first example is used.

Through the electrode equipments, a direct current of 4.5 mA passes in a liquid to be treated in the container 1; and in the case that the direct current is of this value, H ions and OH ions generate as shown in the following Chemical Formula 8, without generating hydrogen gas and oxygen gas.



The OH ions and the hydroxyl group in the adsorbent 5 bind to the heavy metal ions in the waste fluid, thereby causing flocking as afore-mentioned; the flocked particles are aggregated by adsorbent 5; and then they are trapped with the second filter layer 4.

In this manner, the heavy metal ions can be removed more effectively by placing the electrode equipments and electrically charging a direct current of an appointed value, thereby generating compulsorily OH ions in the waste fluid.

Although in the above-described example, a case that a direct voltage was charged between the electrode 6 and the container 1 made of conductor was shown, in an alternative embodiment an earth electrode or a negative electrode may be equipped separately from the container 1.

In addition, as a further alternative embodiment, the electrode 6 may be omitted by replacing the first filter layer 3 consisting of a filter paper of 100  $\mu\text{m}$  by a network

electrode of 100  $\mu$ m mesh stainless steel.

Moreover, although the above-described example has shown a case that a direct voltage of +2 V was charged, the important thing is not the voltage value but is an electric current value for generating OH ions shown in Chemical Formula 8.

That is, in the apparatus in Fig. 6, when the electric current value passed through water exceeds about 9.2 mA, oxygen gas is generated, and when it exceeds about 23 mA, hydrogen gas is generated.

Therefore, by taking a safety factor of twice, preferred electric current value is about 4.5 mA; on the other hand, allowable maximum electric current value is about 23 mA, above which hydrogen gas is generated, or under.

[Third Example]

Fig. 7 is a side sectional view showing a third example of the invention. The difference of the third example from the example in Fig. 6 is that powders of cellulose are used as the adsorbent 5.

That is, while the second example in Fig. 6 has used an adsorbent having an aggregating ability and hydroxyl group as shown in [Chemical Formula 1], [Chemical Formula 2] and [Chemical formula 3], the example in Fig. 7 has used an adsorbent having no hydroxyl group. As the adsorbent 5 in Fig. 7, powders of cellulose acting to assist filtration by the second filter layer 4 was used; OH ions

for binding to the heavy metal ions depends exclusively on that given by a network electrode 6.

In the third example in Fig. 7, the first filter layer 3 in Fig. 6 is changed to a network electrode 6, similarly to the modified example described in the latter half of the second example of Fig. 6.

Next, description is made on an example for removing heavy metal ions in a silicon electrolysis waste fluid with the example in Fig. 7.

Fig. 8, Fig. 9 and Fig. 10 are graphs showing a condition of metal contents obtained by an X-ray microanalyzer similar to those in Fig. 3, Fig. 4 and Fig. 5. Fig. 8 shows metal contents in a waste fluid from silicon electrolysis before purification; Fig. 9 shows metal contents in the purification liquid after 3 times of purification frequency purifying with a cartridge filter of 10  $\mu$ m as the second filter layer 4, and Fig. 10 shows metal contents in the purified liquid after 20 times of repeated circulation purification frequency purifying with a cartridge filter of 10  $\mu$ m as the second filter layer 4, respectively.

In addition, data thereof are summarized in Table 5.

[Table 5]

Metal	Co	Fe	Cu	Si
Waste fluid (mg/l)	7.76	5.20	9.08	30.17
3 times (mg/l)	0.20	0.84	5.29	0.00
20 times (mg/l)	0.11	1.11	5.25	0.00

As shown in Table 5, the heavy metals were removed to an almost satisfactory level in the example in Fig. 7, too.

Fig. 11 and Fig. 12 are magnified photographs in 30 magnifications and 300 magnifications obtained by a scanning electronic microscope, respectively, of an adsorbent which is composed of 3 substances shown by [Chemical Formula 1], [Chemical Formula 2] and [Chemical formula3]. According to Fig. 11, the adsorbent is of minute particle form, but each of the minute particles further has a structure in which minute needles gathered in the pill-like form according to 300 times magnification photograph in Fig. 12. It is estimated from the phenomenological theory that such a minute structure closely relates to the effective removal of the minute particles in the liquid to be treated having far smaller size than the spaces among the minute particle forms.

### INDUSTRIAL APPLICABILITY

As described above, according to the invention, the minute particles in the liquid to be purified can be removed without use of a large amount of an aggregation agent as in the conventional method, and with use of a filter having large openings about 100 times as coarse as the size of the minute particles to be removed; and moreover, the metal ions or the heavy metal ions in the liquid to be treated can be removed without using a chelate agent or an aggregation agent; and therefore, the invention has a great industrial applicability.

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AMENDMENT UNDER ARTICLE 19

CLAIMS

1. (deleted)

2. A purification apparatus for a liquid to be treated characterized in that it has first and second filter layers for filtering the liquid to be treated,

and an adsorbent placed between said first and second filter layers and containing at least basic magnesium sulfate and magnesium hydroxide,

and in that it is designed such that, by passing a liquid to be treated containing metal ions through said adsorbent, the metal ions in the liquid to be treated is bound to a hydroxyl group in an adsorbent resulting in flocculation, flocculated particles are aggregated so that a mass is formed, and then trapped by said second filter layer.

3. A purification apparatus for a liquid to be treated characterized in that it has first and second filter layers for filtering the liquid to be treated,

an adsorbent placed between said first and second filter layers and containing at least basic magnesium sulfate and magnesium hydroxide,



and electrode equipments for electrically charging a direct current through said liquid to be treated and generating OH ions in the liquid,

and in that it is designed such that the OH ions generated by said electrode equipments as well as the hydroxyl group in said adsorbent are bound to the metal ions in the liquid to be treated resulting in flocculation, particles flocculated in this manner are aggregated, and then trapped by said second filter layer.

4. A purification apparatus for a liquid to be treated characterized in that it has first and second filter layers for filtering the liquid to be treated,

an adsorbent placed between said first and second filter layers and containing a powdery cellulose,

and electrode equipments for electrically charging a direct current through said liquid to be treated and generating OH ions in the liquid to be treated,

and in that it is designed such that the OH ions generated by said electrode equipments is bound to the metal ions in the liquid to be treated resulting in flocculation, particles flocculated in this manner are trapped by said adsorbent and said second filter layer.

5. (amended) The apparatus according to anyone of claims 2 to 4, wherein said metal ions are heavy metal

ions.

6. (deleted)

7. A process for purifying a liquid to be treated characterized in that it comprises:

step in which the liquid to be treated is passed through a first filter layer,

step in which the liquid to be treated passed through said first filter layer is passed through a layer of an adsorbent placed downstream of said first filter layer, subsequently thereto, and comprising at least basic magnesium sulfate and magnesium hydroxide,

step in which said metal ions is bound by further passing the liquid to be treated passed through the layer of adsorbent through a layer of the second filter layer placed downstream of said layer of adsorbent and subsequently thereto causing flocculation, and further aggregating the flocculated particles so that a mass is formed, and

step in which the liquid to be treated passed through said layer of adsorbent is trapped by a second filter layer placed downstream of said layer of adsorbent and subsequently thereto.

8. A process for purifying a liquid to be treated

characterized in that it comprises:

step in which the liquid to be treated is passed through a first filter layer,

step in which a direct current is electrically charged through said liquid to be treated so that OH ions is generated in the liquid to be treated,

step in which the liquid to be treated passed through said first filter layer is passed through a layer of an adsorbent placed downstream of said first filter layer, subsequently thereto, and containing at least basic magnesium sulfate and magnesium hydroxide,

step in which the OH ions generated by said electric current as well as a hydroxyl group in said adsorbent are bound to the metal ions in the liquid to be treated resulting in flocculation, and further flocculated particles are aggregated, and

step in which an aggregate formed in said aggregation is trapped by a second filter layer placed downstream of said layer of adsorbent and subsequently thereto.

9. A process for purifying a liquid to be treated characterized in that it comprises:

step in which the liquid to be treated is passed through a first filter layer,

step in which a direct current is electrically

charged through said liquid to be treated so that OH ions is generated in the liquid to be treated,

step in which the liquid to be treated passed through said first filter layer is passed through a layer of an adsorbent placed downstream of said first filter layer, subsequently thereto, and containing powdery cellulose,

step in which the OH ions generated by said electric current is bound to the metal ions in the liquid resulting in flocculation , and

step in which particles flocculated in this manner are trapped by said layer of adsorbent and a second filter layer placed downstream of said layer of adsorbent and subsequently thereto.

10. (amended) The process according to anyone of claims 7 to 9, wherein said metal ions is a heavy metal ions.

BRIEF DESCRIPTION UNDER ARTICLE 19, PARAGRAPH (1), PCT

[1] Because a paragraph [Constitution] on page 1 of JP-A-6-226017, cited in the International Search Report, discloses "a granular or powdery adsorbent 3 of magnesium is placed between a first cylindrical body 1 and a second cylindrical body 2", the inventive step of claims 1 and 6 is greatly affected. Therefore, claims 1 and 6 are deleted.

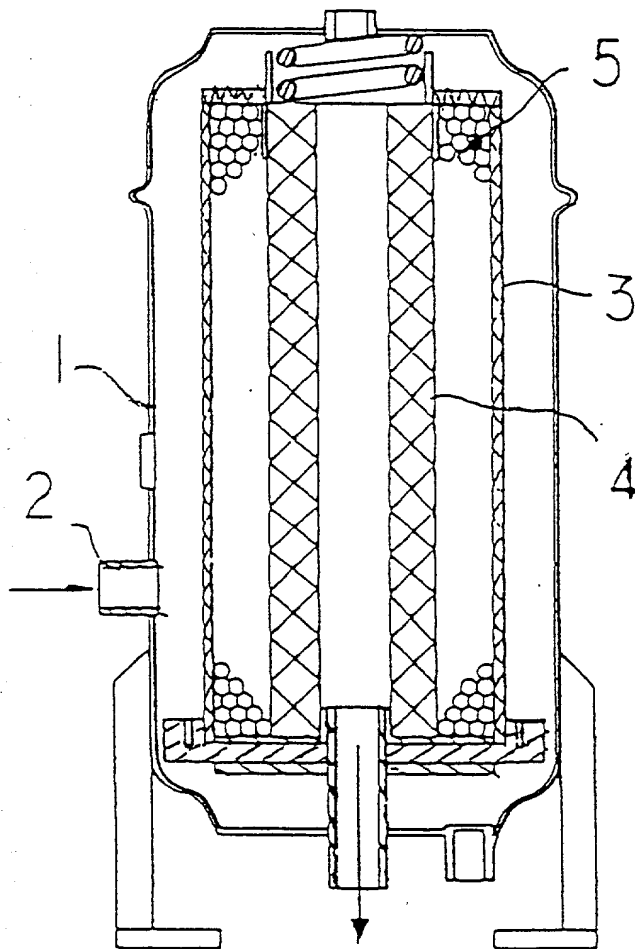
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Abstract

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A clarification apparatus for liquid, characterized in that it has a first filtration layer (3) and a second filtration layer (4) and, arranged between the layers, an absorbing agent (5) comprising basic magnesium sulfate and magnesium hydroxide, wherein a liquid to be treated containing fine particles is passed through the absorbing agent (5), to thereby aggregate the fine particles in the liquid to be treated, and then the resulting large particles are caught by the second filtration layer (4) and wherein a liquid to be treated containing a heavy metal ion is passed through the absorbing agent (5), to thereby react the metal ion with a hydroxyl ion of the absorbing agent and solidify the metal ion, followed by aggregation, and then the resulting large particles are caught by the second filtration layer (4); and an apparatus which further has an electrode device capable of generating an OH ion, wherein the electrically generated OH ions, the OH radicals of the absorbing agent (5) and the heavy metal ions react with one another, to thereby efficiently solidify the metal ions and aggregate the resultant solid into large particles, and then the resulting large particles are caught by the second filtration layer (4).

FIG. 1



- 1: Container
- 2: Inlet for waste fluid
- 3: First filter layer
- 4: Second filter layer
- 5: Adsorbent

FIG. 2

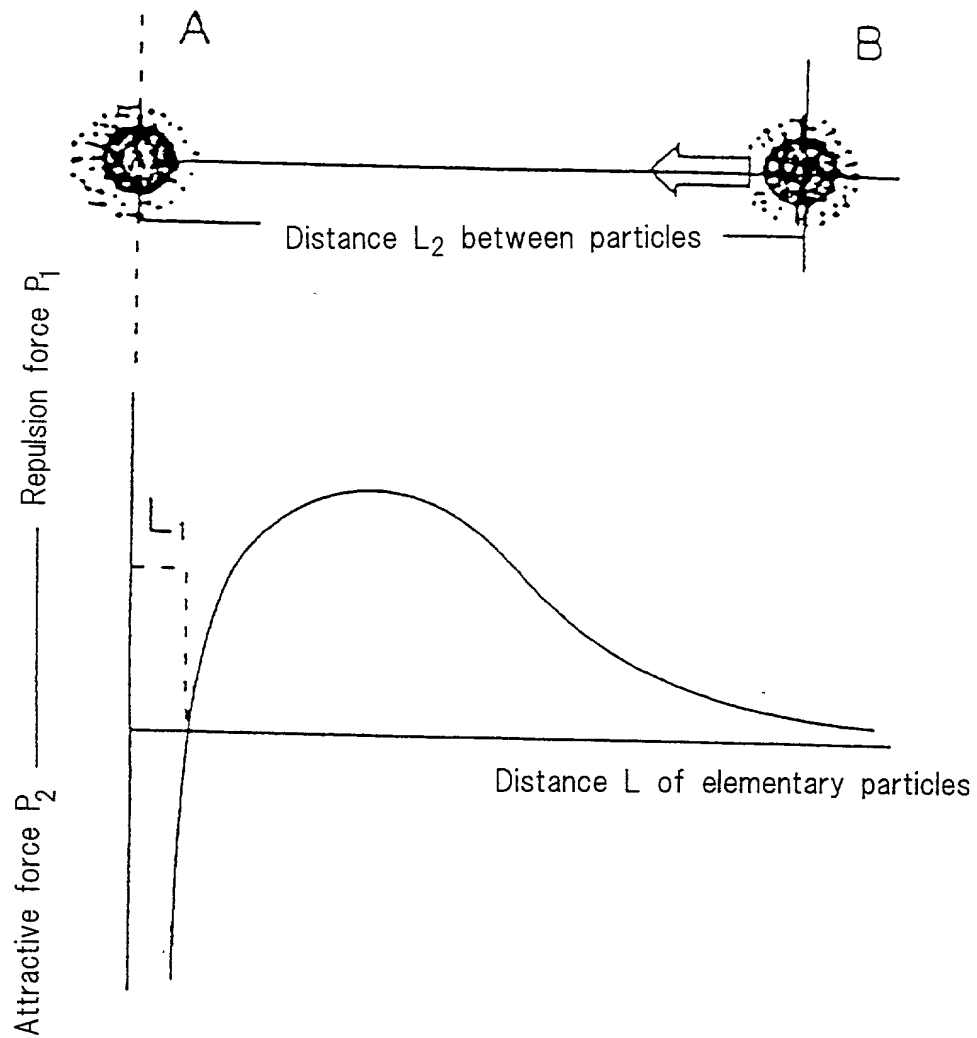




FIG. 3

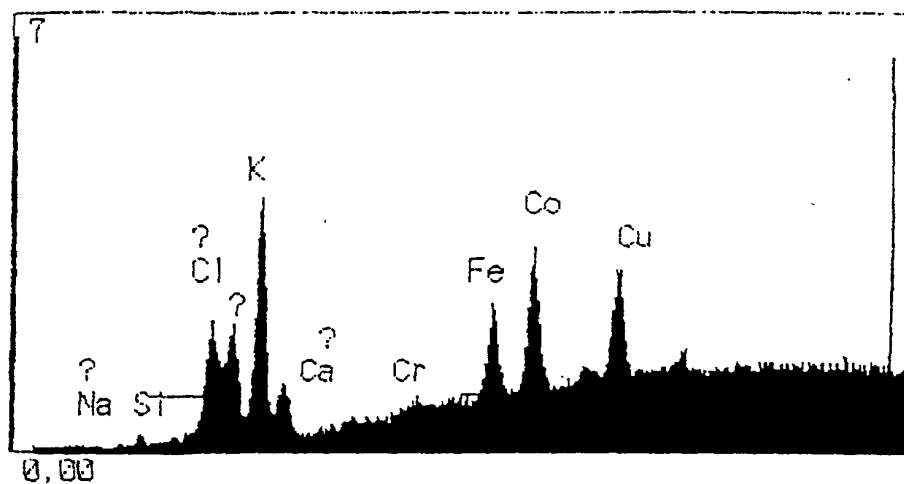


FIG. 4

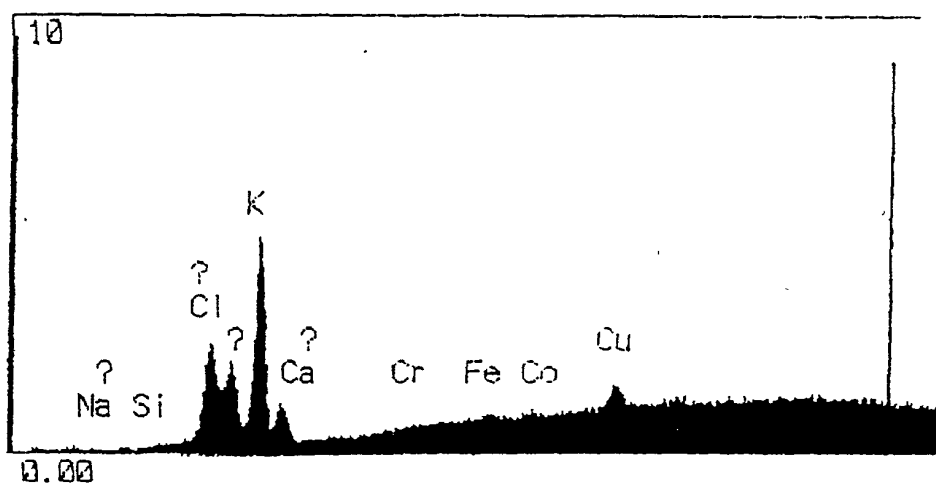
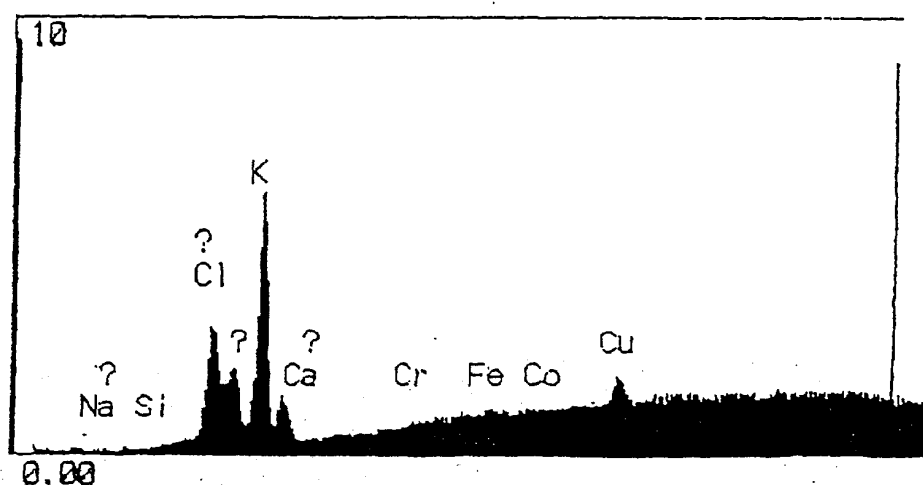


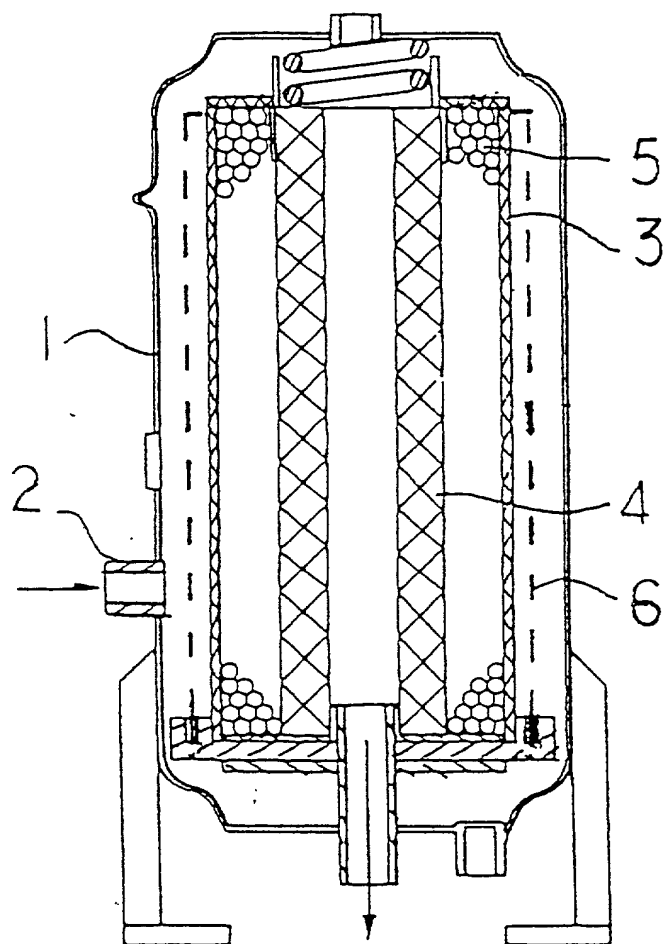
FIG. 5



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FIG. 6



6: Electrode

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FIG. 7

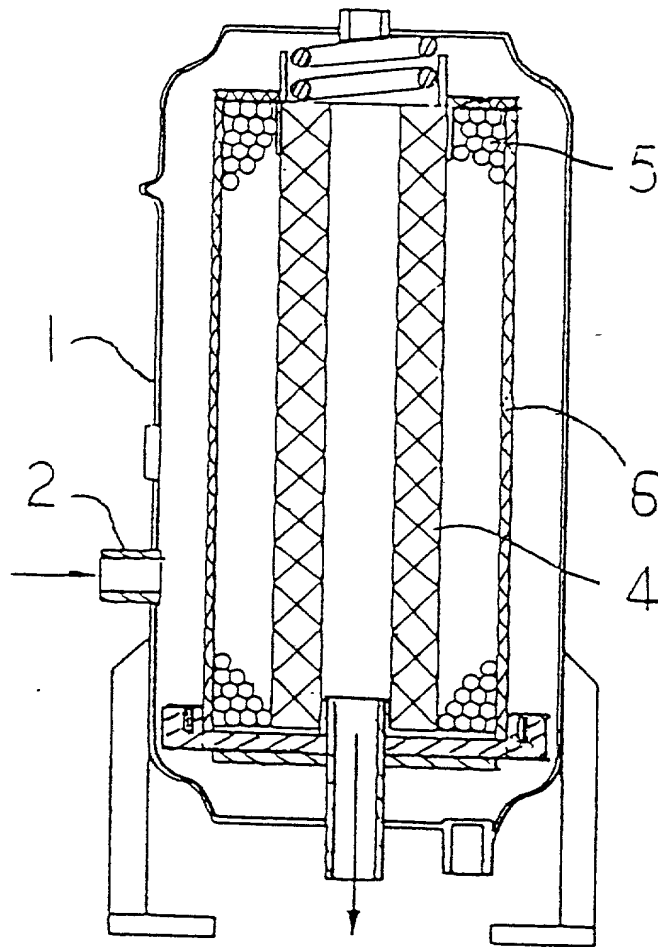


FIG. 8

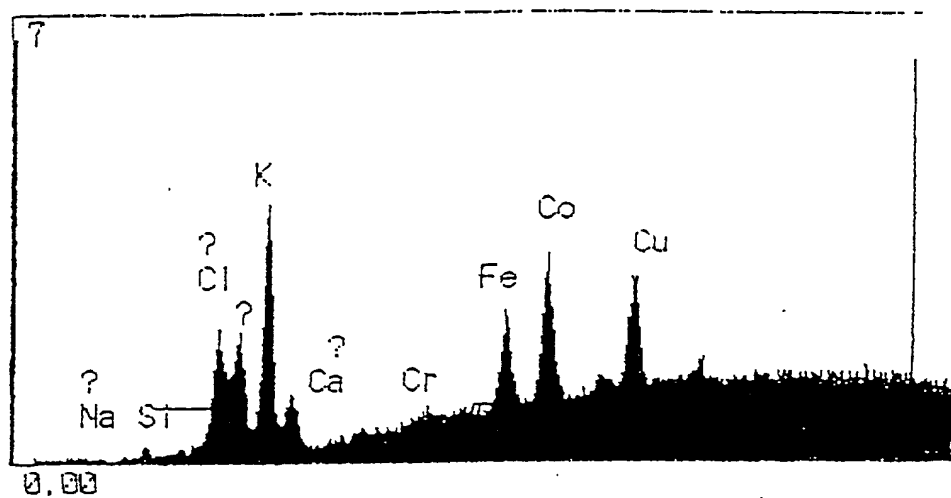


FIG. 9

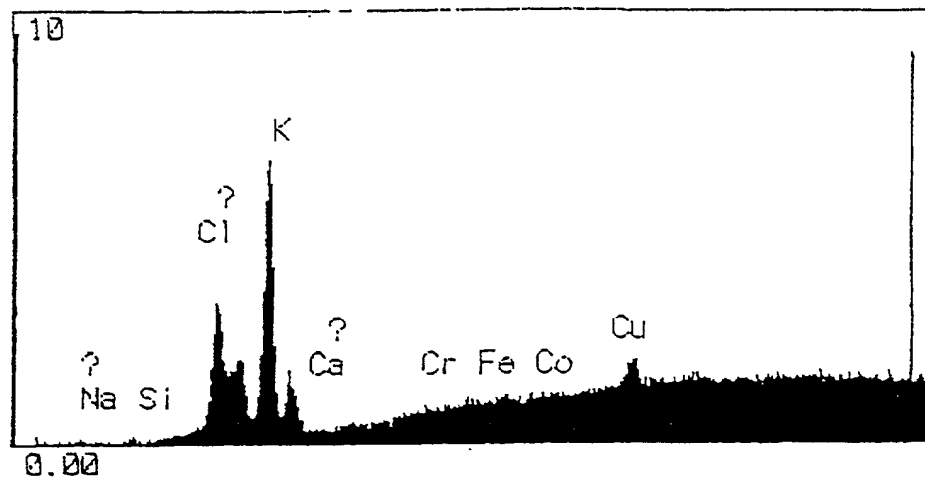
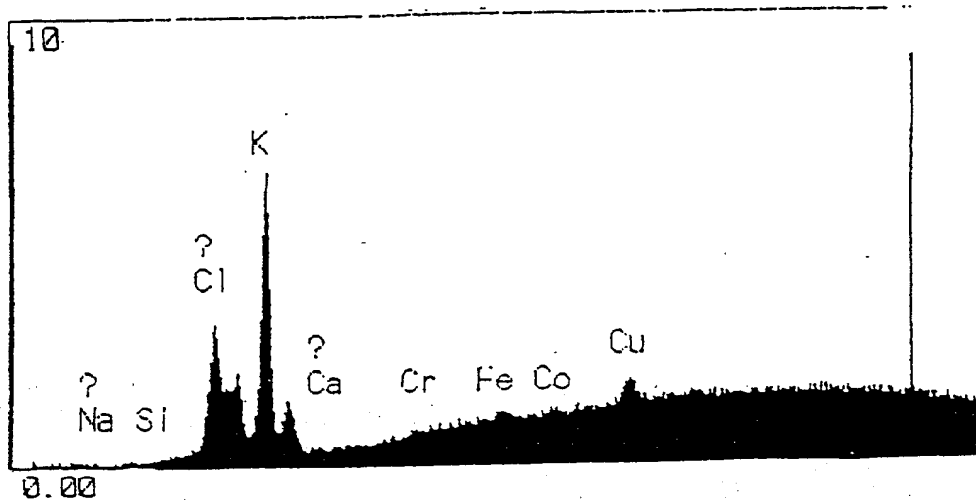


FIG. 10



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FIG. 11

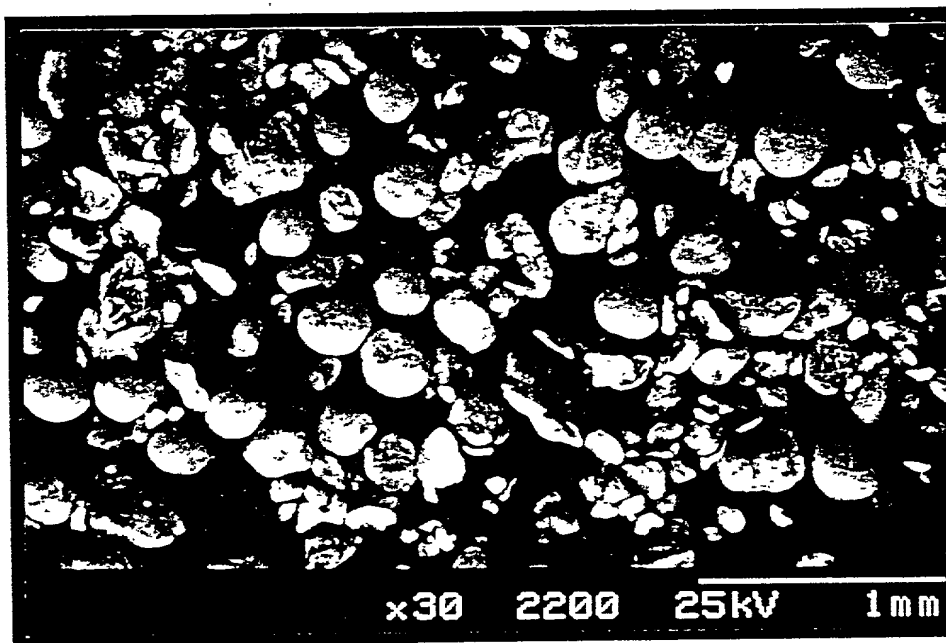
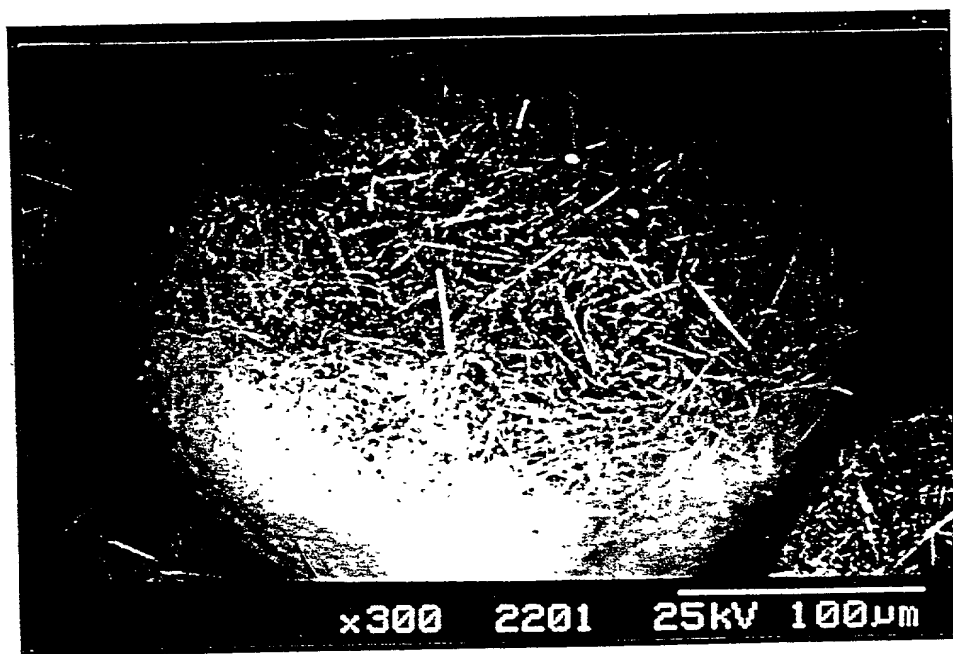


FIG. 12



## Declaration and Power of Attorney For Patent Application

特許出願宣言書及び委任状

Japanese Language Declaration

日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。

私の住所、私書箱、国籍は下記の私の氏名の後に記載された通りです。

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者（下記の名称が複数の場合）であると信じています：

上記発明の明細書は、

☐ 本書に添付されています。

☐ 月 日に提出され、米国出願番号または特許協定条約国際出願番号を とし、（該当する場合）に訂正されました。

私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容を理解していることをここに表明します。

私は、連邦規則法典第37編第1条56項に定義されるとおり、特許資格の有無について重要な情報を開示する義務があることを認めます。

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

LIQUID PURIFICATION APPARATUS

the specification of which

☐ is attached hereto.

☒ was filed on November 29, 2000 as United States Application Number or PCT International Application Number PCT/JP00/08458 and was amended on (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

## Japanese Language Declaration

(日本語宣言書)

私は、米国法典第35編119条(a)-(d)項又は365条(b)項に基づき下記の、米国以外の国の少なくとも一カ国を指定している特許協力条約365(a)項に基づく国際出願、又は外国での特許出願もしくは発明者証の出願についての外国優先権をここに主張するとともに、優先権を主張している、本出願の前に出願された特許または発明者証の外国出願を以下に、枠内をマークすることで、示しています。

## Prior Foreign Application(s)

外国での先行出願

(Number) (番号)	(Country) (国名)
Pat. Hei 11-376283	Japan

私は、第35編米国法典119条(e)項に基づいて下記の米国特許出願規定に記載された権利をここに主張いたします。

(Application No.) (出願番号)	(Filing Date) (出願日)

私は、下記の米国法典第35編120条に基づいて下記の米国特許出願に記載された権利、又は米国を指定している特許協力条約365条(c)に基づく権利をここに主張します。また、本出願の各請求範囲の内容が米国法典第35編112条第1項又は特許協力条約で規定された方法で先行する米国特許出願に開示されていない限り、その先行米国出願書提出日以降で本出願書の日本国内または特許協力条約国際提出日までの期間中に入手された、連邦規則法典第37編1条56項で定義された特許資格の有無に関する重要な情報について開示義務があることを認識しています。

I hereby claim foreign priority under Title 35, United States Code, Section 119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Priority Claimed

優先権主張

Yes No

(Day/Month/Year Filed) (出願年月日)	はい	いいえ
3 December, 1999	<input checked="" type="checkbox"/>	<input type="checkbox"/>
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I hereby claim the benefit under Title 35, United States Code, Section 119(e) of any United States provisional application(s) listed below.

(Application No.) (出願番号)	(Filing Date) (出願日)

I hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code Section 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of application.

(Application No.)

(Filing Date)

(Status : Patented, Pending, Abandoned) (現況 : 特許許可済、係属中、放棄済)

(Application No.)

(Filing Date)

(Status : Patented, Pending, Abandoned)

私は、私自身の知識に基づいて本宣言書中で私が行う表明が真実であり、かつ私の入手した情報と私の信じるところに基づく表明が全て真実であると信じていること、さらに故意になされた虚偽の表明及びそれと同等の行為は米国法典第18編第1001条に基づき、罰金または拘禁、もしくはその両方により処罰されること、そしてそのような故意による虚偽の声明を行えば、出願した、又は既に許可された特許の有効性が失われることを認識し、よってここに上記のごとく宣誓を致します。

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

委任状：私は下記発明者として、以下の代理人をここに選任し、本願の手続を遂行すること並びにこれに関する一切の行為の特許商標庁に対して行うことを委任する。(代理人氏名および登録番号を明記のこと)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. (list name and registration number)

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発明者の署名	日付	Inventor's signature <u>Kimihiko Okaue</u> Date July 19, 2001
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国籍	Citizenship Japanese	
郵便の宛先	Post Office Address 6-11, Hoshoen, Takarazuka-shi, Hyogo 655-0016 Japan	

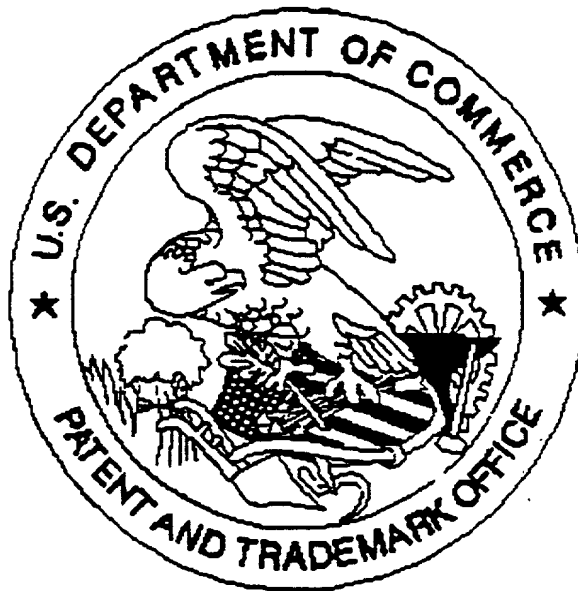
第二共同発明者	Full name of second joint inventor, if any	
第二共同発明者の署名	日付	Second inventor's signature Date
住所	Residence	
国籍	Citizenship	
郵便の宛先	Post Office Address	

第三共同発明者	Full name of third joint inventor, if any	
第三共同発明者の署名	日付	Third inventor's signature Date
住所	Residence	
国籍	Citizenship	
郵便の宛先	Post Office Address	

(第四またはそれ以降の共同発明者に対しても同様な情報および署名を提供すること。)

(Supply similar information and signature for fourth and subsequent joint inventors.)

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